Integral cross sections for electron-impact excitation of the 4 ^2P state in copper

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We report integral cross sections for electron-impact excitation of the 4 ^2P state in copper for incident electrons with energies in the range from threshold to 100 eV. Measurements, based on an optical excitation function procedure, are compared with coupled-channel and coupled-channel-optical method calculations that we have also performed as a part of this study. Agreement between our measurements and theory was generally only modest. The present measurements are also found to be in quite good accord with the early near-threshold integral cross sections of Flynn *et al.* [C. Flynn, Z. Wei, and B. Stumpf, Phys. Rev. A **48**, 1239 (1993)] and the higher energy measurements from Ismail and Teubner [M. Ismail and P. J. O. Teubner, J. Phys. B **28**, 4149 (1995)]. Where possible, comparison of our data is also made with earlier theory.

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I. INTRODUCTION

Lasers have found major application in technologies including communications, remote sensing, medicine and active imaging. In particular gas discharge lasers, operating on transitions from resonance to metastable levels, are characterized by high efficiency and high average power so that with their low running costs they are ideal for many industrial applications. One such laser of this type, the copper vapor laser (CVL), is now well established as a very useful source of high-power visible light [1].

The performance of the CVL and its further development depend on how adequate the corresponding kinetic models for the process are [2]. The adequacy of these models in turn depends crucially on the accuracy of the parameters that are included. For example, reliable values in the modeling are needed for quantities such as lifetimes, transition probabilities, bound-state energies including fine and hyperfine structure splittings, and the appropriate cross sections for the various excitation processes. Low-energy electron-impact excitation of the 4 ^2P state is the dominant process for population inversion in the CVL. Unlike the excitation mechanism for the 4 ^2P state, electron-impact excitation of the metastable 3 ^2D state is weak since the transition 4 $^2S \rightarrow 3\ ^2D$ is optically forbidden on parity grounds. Strong radiation trapping at 1550 °C inhibits the decay of the 4 ^2P excited state directly back into ground state, via the emission of 325 and 327 nm radiation, thus effectively creating a three-level system for pulsed-laser operation. The electron excitation of the 4 ^2P and 3 ^2D states in copper is clearly very relevant to an understanding of the CVL and the cross sections for these excitation processes are among the most important parameters in modeling the CVL [3]. This has historically been the primary motivation for theoretical and experimental interest in electron-copper scattering and, at least in part, also was an important rationale in our present 4 ^2P state study.

There have been several theoretical approaches used to study the electron-impact excitation of the 4 ^2P state in Cu. Winter [4], Peterkop and Liepinsh [5], and Sobelman *et al.* [6] used the Born approximation with different descriptions of the target wave functions. Provided the target wave functions are physical, as evidenced by them giving good agreement with the known 4 $^2S \rightarrow 4\ ^2P$ optical oscillator strength [7–9], the Born approximation should provide a reasonable description of the scattering process for energies greater than about 100 eV [10]. A semiclassical impact-parameter calculation was performed by Winter and Hazi [11] to give integral cross sections (ICSs) for excitation of the 4 ^2P state by electrons with energies in the range of 3.8–200 eV. More advanced first-order distorted-wave approximation (DWA) calculations were undertaken by Pangantiwar and Srivastava [12]. This was for a nonrelativistic formulation, which was later extended to a relativistic description by Srivastava *et al.* [13] for energies in the range of 20–100 eV. A second-order nonrelativistic DWA calculation for 4 $^2S \rightarrow 4\ ^2P$ excitation was subsequently reported by Madison *et al.* [14], in this case for incident electrons with energies between 10–100 eV. Another theoretical approach that has been used to study this excitation process in copper is the coupled-channel (CC) methodology. Within this paradigm we note the four-state (4*s*, 3*d*, 4*p*, and 4*d*) CC results from Msezane and Henry [15] and Scheibner *et al.* [16]. Scheibner and Hazi apparently later extended their model to ten states (see Flynn *et al.* [17]), but this work was never independently published. Fi

nally, we note the coupled-channel-optical (CCO) method and convergent close-coupling (CCC) method 4 ^2P results from Zhou *et al.* [18]. Both these methods reported ICS for energies in the range of 20–100 eV, with the CCO including eight states in their *P*-space expansion [18] while the CCC calculations used expansions that ranged from 50 to 65 states. Very recently, Maslov *et al.* [19] employed a fully relativistic *B*-spline *R*-matrix method for electron-impact excitation of the $^2S_{1/2} \rightarrow {}^2P_{1/2,3/2}$ resonance transitions in gold. It would be very interesting if that approach was also applied to the resonance transition in copper.

An experimental optical excitation function method, in which the ~325 and ~327 nm photons arising from the decay of the 4 ^2P state to the ground state are observed, was used in three previous crossed-beam experiments to measure ICSs for the 4 ^2P state excitation. In the case of Borozdin *et al.* [20] the measured cross sections, in the energy range of 10–200 eV, were made absolute by comparing the radiation with lines of nitrogen which was present as a residual gas of known density. The ICSs reported by Aleksakhin *et al.* [21] were put on an absolute scale by comparing the 325–327 nm radiation with that of a standard ribbon-filament lamp. Those measurements were made in the energy range of 10–75 eV. As neither the data of Borozdin *et al.* [20] nor those of Aleksakhin *et al.* [21] are now considered reliable, we do not discuss them further here. Flynn *et al.* [17] normalized their excitation function to the Born integral cross section at 1000 eV and made estimates for the contributions of cascades to the observed signal. One of our motivations for the present ICS measurements was that Flynn *et al.* limited their published measurements to the 3.8–8 eV energy range so that those data not only need to be cross checked but also need to be extended. ICS can also be derived from differential cross-section (DCS) data. This approach was originally utilized by Trajmar *et al.* [22], but as noted by both Msezane and Henry [15] and Yousif Al-Mulla [23] those data are unreliable, having to be scaled by an energy-independent factor of 0.36. That factor was obtained by comparing calculated generalized oscillator strengths with those deduced from the angular distribution measurements of Trajmar *et al.* [22]. More recently, Ismail and Teubner [10] reported ICS from this approach for electrons with incident energies of 20, 40, 60, 80, and 100 eV. Hence, another aim of this work is to report ICS on a finer energy grid than is currently known. This can be particularly important if resonances play a role in the scattering process [15].

In the next section we describe our apparatus and measurement techniques, including our normalization procedure. In Sec. III details of our coupled-channel and coupled-channel-optical-model calculations are provided before our results and a discussion of those results are detailed. Finally, in Sec. V, some conclusions from the present study will be drawn.

II. EXPERIMENTAL DETAILS

The excitation function for the *P* states was measured by observing photons emitted in the decay of the $(3d^{10}4p)^2P_{1/2}$ and $(3d^{10}4p)^2P_{3/2}$ states with a photomultiplier tube equipped with an interference filter. The filter had a bandpass of 10 nm centered at 330 nm, which enabled both the 325 and 327 nm lines to be detected. We note, subject to the caveats discussed below, this excitation function is proportional to the integral cross section (Q) of interest. The detected photons arose from the interaction region that was defined by the intersection of a well-focused electron beam and a beam of copper atoms. This region acted as the focal point of the objective lens of the optical system which was oriented at an angle of 54°44′ with respect to the electron beam. Thus our measurements were not influenced by any polarization effects.

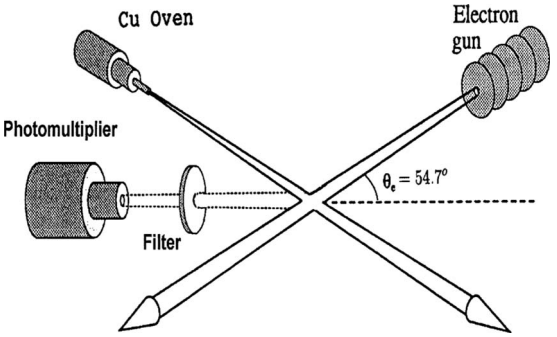

FIG. 1. Schematic diagram of the experimental geometry for the present optical excitation function measurements.

A schematic diagram of our experimental configuration is given in Fig. 1. Pulses from the photomultiplier tube were amplified and counted in either of two gated scalers. The gates were provided by chopping the electron beam.

The electron gun produced currents between 0.5 and 30 μA over the energy range of the experiments. The current was collected in a Faraday cup which was monitored by a computer. It was established that the photon signal was proportional to electron-beam current.

The copper atoms were produced by heating copper wire in a molybdenum oven that was heated by electron bombardment. Typical oven temperatures around 1550 K produced a copper beam which had a diameter of 6.5 mm and an angular divergence of 0.1 radians. The oven temperature was stable to within 2 K over the course of a run which gave rise to a maximum correction in the beam density of 3% at 1550 K.

The electron-beam profile was not constant over the energy range of the experiments. Measurements showed that the electron-beam diameter changed from 4 mm at 100 eV to 5 mm at 5 eV. To first order these changes did not affect the detected photon signal because the electron-beam diameter was always less than the diameter of the copper beam. The residual influence on the solid angle of the photon detection system was small because the radius of the electron beam was small compared to the focal length of the objective lens.

During the course of the experiments we investigated the possible influence of $3d^9 4s^2\ ^2D$ states on the incident copper beam. These states were detected by tuning a laser beam to the $^2D_{3/2}$-$^2P_{1/2}$ transition at 578 nm and observing the fluorescence at 327 nm. The origin of these *D* states in the beam was shown to arise from the Boltzmann distribution of states in the oven. The density of *D* states in the initial beam was about 5 orders of magnitude less than the ground-state copper atom density.

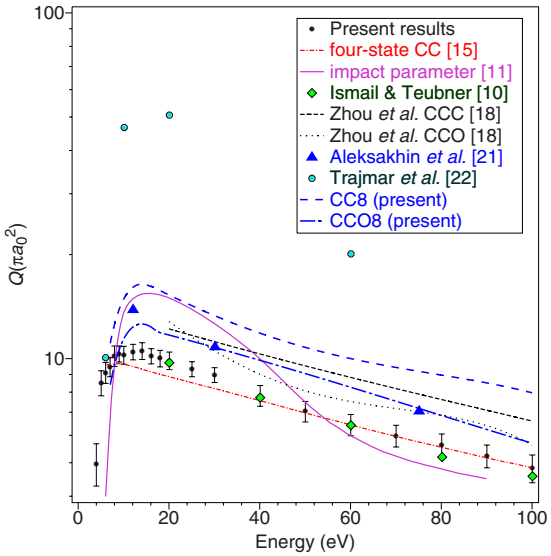

FIG. 2. (Color online) Integral cross sections (πa_0^2) for electron-impact excitation of the 4 ^2P state of copper for the present experiments (●) our CCO8 (—— –) and CC8 (— —) computations and for previous work as labeled in the legend.

In principle the excitation function contains cascade contributions from levels higher than the P states. The problems associated with assigning these contributions have been discussed by Suvorov [24]. With respect to the normalization of our excitation function to the calculation of Msezane and Henry [15] at 100 eV, there is good evidence to suspect that cascades have no effect at 100 eV. The cross sections of Ismail and Teubner [10] were obtained by integrating the differential cross sections for the excitation of the P states at each energy. They are free from cascades. We note the excellent agreement between our normalized cross sections and those of Ismail and Teubner even down to 20 eV. This comparison is shown in Fig. 2. The influence of cascades at lower energies however cannot be ignored, although they can in principle be corrected for [19].

III. THEORETICAL METHODOLOGY

A comprehensive review of the application of the coupled-channel-optical method to electron-atom scattering can be found in McCarthy and Weigold [25], with a nice summary of that work for electron-copper scattering being found in Zhou *et al.* [18]. As a consequence, for the sake of completeness, only a brief precis of the more important details of our CCO method calculation is given here.

The transition from a target state j to a state i is calculated in terms of the set of coupled Lippmann-Schwinger integral equations

$$\langle \boldsymbol{k}_i i|T|j\boldsymbol{k}_j\rangle = \langle \boldsymbol{k}_i i|V|j\boldsymbol{k}_j\rangle + \sum_l \int d^3k \langle \boldsymbol{k}_i i|V|l\boldsymbol{k}\rangle \frac{1}{E^{(+)} - \epsilon_l - \frac{1}{2}k^2} \langle \boldsymbol{k}l|T|j\boldsymbol{k}_j\rangle, \tag{1}$$

where the complete set l includes bound and continuum target states and the sum over l implies integration for the continuum. Momenta of the external electrons are denoted by $\boldsymbol{k}$. The T matrix for the transition is T and the potential experienced by the external electron is V. Matrix elements of T and V are antisymmetrized.

The CCO method divides the target space into two complementary spaces P and Q. P space includes the observed states and other discrete states that are strongly coupled enough to affect the result. Q space includes the target continuum. The set l of target states used to expand the Green's function in the Lippmann–Schwinger equation is truncated to P space. The effect of Q space is included by adding a complex, nonlocal polarization potential W^Q to V, resulting in the optical potential V^Q which is formally defined by

$$\langle \boldsymbol{k}' i|V^Q|j\boldsymbol{k}\rangle = \langle \boldsymbol{k}' i|V + VQ\frac{1}{Q(E^{(+)} - H_t - V - K)Q}QV|j\boldsymbol{k}\rangle, \tag{2}$$

where K is the kinetic-energy operator for the external electron, H_t is the Hamiltonian of the target and Q is the projection operator for Q space. All amplitudes are antisymmetrized.

In implementing the CCO method P space includes enough discrete channels for convergence and Q space is only the continuum, whose momentum-space representation is

$$|\Psi_l^{(-)}(\boldsymbol{q})\rangle = |\boldsymbol{p}\,\psi^{(-)}(\boldsymbol{q})\rangle, \tag{3}$$

where $|\psi^{(-)}(\boldsymbol{q})\rangle$ is a time-reversed Coulomb function, orthogonalized to the appropriate target orbital in i or j, and $\boldsymbol{p}$ and $\boldsymbol{q}$ are, respectively, the momenta of the faster and slower continuum electrons. This simplification is necessary because of the six-dimensional momentum integration, which is performed by a multidimensional Cartesian method. The equivalent-local approximation to W^Q is

$$W_{il}^Q(\boldsymbol{k}', \boldsymbol{P}) = \int d\hat{\boldsymbol{P}} \langle \boldsymbol{k}' i|W|l\boldsymbol{k}\rangle, \tag{4}$$

where

$$\boldsymbol{P} = \boldsymbol{k}' - \boldsymbol{k}. \tag{5}$$

The scattering amplitude at a given energy for a local potential depends only on the absolute value of the momentum transfer, with the equivalent-local approximations being expected to improve as the total energy is increased [25].

A simplification of the CCO is where the optical potential is ignored, so that the basis consists only of those states in P space. This approach is known as the coupled-channel methodology [25], and here we extend the earlier four-state CC results from Msezane and Henry [15] and Scheibner *et al.* [16] to eight states $(4s, 4p, 4d, 5s, 5p, 5d, 6s, 6p)$, with all these states being computed within a Hartree-Fock representation.

The present calculations adopt the same structure approximation as Zhou *et al.* [18], namely, copper is treated as a single-electron atom with a closed-shell core. They omit core-excited channels, expecting them to be weakly coupled

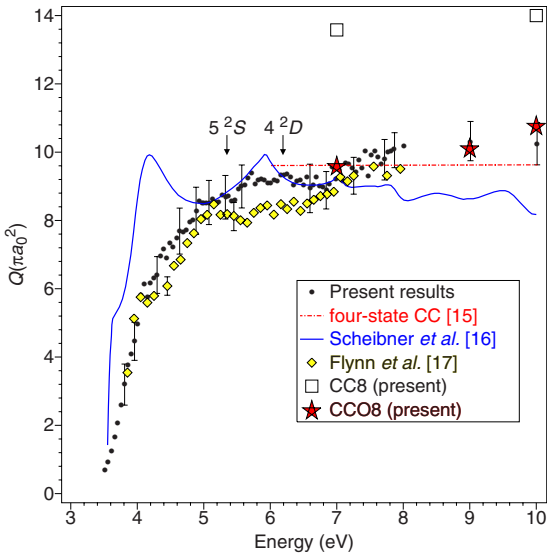

FIG. 3. (Color online) Low-energy integral cross sections (πa_0^2) for electron-impact excitation of the 4 ^2P state of copper for the present experiments (●) our CCO8 (⋆) and CC8 (□) computations and for previous work as labeled in the legend.

to those included in P space. The validity of that assumption is explored in a little more detail in Zhou *et al.* [18].

Our wave functions were computed using the code of Froese-Fischer and Saxena [26]. Although this is in general a multiconfiguration Hartree-Fock (MCHF) architecture, the option for single-channel Hartree-Fock (SCHF) calculations is available and was utilized for the eight P-space states of this work. This approach is thus very similar to that adopted for the copper atomic structure by Zhou *et al.* [18]. Of particular importance to this work was the $4s$-$4p$ transition length and velocity oscillator strength (f) values, which were 1.59 and 1.05, respectively. Clearly these values are far too large for what is to first order a "one-electron" system. Upon comparison of these results to the corresponding accepted experimental value [7–9] with $f \sim 0.64$, we see that our description of the target is thus only fair. Certainly the available MCHF oscillator strength results from Msezane and Henry [15] and Froese-Fischer [27] are in better accord with the experiment than the present, as you might expect.

However, we note that Zhou *et al.* [18] had previously demonstrated that incorporation of Q-space coupling into an optical-model scattering calculation to some extent ameliorates a marginal structure representation. Thus we still expect our calculations to have physical value and to shed light on the scattering dynamics.

IV. RESULTS AND DISCUSSION

The present experimental results, for the energy range from threshold to 100 eV, are plotted in Figs. 2 and 3 and a selection of those cross sections is tabulated in Table I. Also included in these figures are our CCO and CC results, but for the energy range of 7–100 eV, as well as relevant data from previous measurements [10,17,21,22] and calculations [11,15,16,18]. Note that our present theoretical cross sections

TABLE I. Selected present experimental electron-impact excitation cross sections for the copper 4 ^2P state.

Energy (eV)	Q (πa_0^2)	Energy (eV)	Q (πa_0^2)
3.81	3.19 ± 0.60	6.93	9.57 ± 0.69
3.95	4.46 ± 0.57	7.03	9.47 ± 0.58
4.05	5.83 ± 0.62	7.13	9.69 ± 0.64
4.20	6.18 ± 0.53	7.22	9.55 ± 0.52
4.30	6.41 ± 0.54	7.37	9.56 ± 0.54
4.44	6.90 ± 0.59	7.47	9.30 ± 0.59
4.54	7.24 ± 0.73	7.61	9.86 ± 0.62
4.64	7.71 ± 0.67	7.71	9.79 ± 0.60
4.73	7.74 ± 0.55	7.81	10.05 ± 0.57
4.88	8.28 ± 0.70	7.86	10.08 ± 0.51
4.98	8.52 ± 0.71	8	10.18 ± 0.70
5.08	8.52 ± 0.64	9	10.33 ± 0.57
5.22	8.55 ± 0.66	10	10.25 ± 0.62
5.32	8.68 ± 0.59	12	10.47 ± 0.55
5.42	8.58 ± 0.50	14	10.54 ± 0.60
5.66	9.27 ± 0.69	16	10.18 ± 0.52
5.71	9.38 ± 0.63	18	10.03 ± 0.56
5.81	9.18 ± 0.63	20	9.88 ± 0.58
5.91	9.17 ± 0.72	25	9.35 ± 0.46
6.00	9.10 ± 0.64	30	8.98 ± 0.44
6.15	9.32 ± 0.59	40	7.83 ± 0.54
6.25	9.36 ± 0.62	50	7.03 ± 0.48
6.34	9.05 ± 0.54	60	6.43 ± 0.46
6.49	9.05 ± 0.63	70	5.99 ± 0.43
6.59	8.95 ± 0.72	80	5.61 ± 0.45
6.69	9.28 ± 0.71	90	5.23 ± 0.40
6.83	8.89 ± 0.55	100	4.87 ± 0.45

were calculated at 15 discrete energies within that range of 7–100 eV, with the curves representing them in Fig. 2 being an interpolation between those points. Further note that the overall errors on the data in Table I arise from our statistical errors combined in quadrature with the uncertainty in the direct cross section used to normalize the measured data at 100 eV. That normalization cross section was taken from Ismail and Teubner [10].

Let us now consider the results embodied in Fig. 2 in more detail. As noted previously, while the results from Trajmar *et al.* [22] were the first ICS measurements for electron-impact excitation of the 4 ^2P state their data were flawed by the normalization method they used. Specifically Trajmar *et al.* normalized their data to the elastic static exchange DCS calculation of Winter [28] at 100 eV and the scattering angle 40°. This early calculation may not be accurate. In addition, Trajmar *et al.* did not measure their 4 ^2P DCS at scattering angles less than 15°. As the ICS at energies greater than about 20 eV are dominated by the DCS contribution for $\theta \leq 15°$ [10], any small error in their extrapolation to 0° could cause a significant error in the ICS they derived. In any event, it is clear from Fig. 2 that the ICSs of Trajmar *et al.*

are generally too large in magnitude. The present experimental results are clearly (Fig. 2) in very good accord with the earlier ICS of Ismail and Teubner [10], which recall were determined from their DCS data and thus do not suffer from any potential cascade contributions. For example, at 20 eV the difference between them is only 1.4%, which suggests that the cascade contribution at this energy is similar to that which was prescribed at 100 eV [24]. On the other hand the results of Aleksakhin *et al.* [21] are not supported by the present measurements. We believe this is due to them having a significant error in the determination of the density of their copper atoms, a point that was previously noted by Msezane and Henry [15].

The predictions of the four-state close-coupling calculation of Msezane and Henry [15] are also shown in Fig. 2, where it is seen that there is good accord with the present measurements for energies greater than 40 eV. This probably reflects, at least in part, the excellent agreement these authors achieved between their length and velocity forms of the optical oscillator strength for the $4s$-$4p$ transition (0.644), and the corresponding experimental values [7–9] of 0.65 ± 0.065 and 0.633, respectively. In the energy range $10 \leq E_0 \leq 30$ eV this close-coupling [15] calculation, however, underestimates the magnitude of the ICS, which would be expected if the cascade contribution was large. However, the excellent agreement between our experimental ICS results and those from Ismail and Teubner [10], as well as the extensive discussion on possible cascade effects in this system by Suvorov [24], indicates that the cascade contribution is smaller than the difference between our measured data and their theory [15]. At energies less than 10 eV the observed agreement in Fig. 2 between our experiment and the four-state CC theory is probably fortuitous because the close-coupling theory does not predict a peak in the cross section at about 15 eV. Also shown in Fig. 2 are the CCC and CCO calculations of Zhou *et al.* [18], where it is clear that their ICS results are uniformly higher in magnitude than our data. In principle both the CCC and CCO calculations should be superior to the four-state CC. However the wave functions employed in Zhou *et al.* [18] were single-configuration Hartree-Fock while those in Msezane and Henry [15] were at the multiconfiguration level. Unfortunately Zhou *et al.* did not publish their oscillator strengths for the $4s$-$4p$ transition, but we strongly suspect they would be inferior compared to those of Msezane and Henry. Hence the somewhat, at first glance anyway, paradoxical CCC and CCO ICS results might simply be a reflection of their less accurate description of the target, compared to Msezane and Henry, in this case.

The present eight-state CC (CC8) and eight-state CCO computations both find the peak in the $4\ ^2P$ integral cross section to occur at around 14.5 eV, in reasonable accord with our experimental result. The magnitude of the current CC8 ICS is, however, typically a factor of ~ 1.6 greater than that of the experiment. Recall that the velocity oscillator strength value for our $4s$-$4p$ transition was calculated to be 1.05, a factor of ~ 1.64 greater than the accepted experimental value [7–9]. This correspondence, we believe, is no coincidence. Rather it suggests that the significant problem with our CC8 calculation is due to the somewhat limited accuracy of our target states. Agreement between our eight-state CCO (CCO8) calculation and our experimental data is better than that achieved with the CC8, although it typically remains, at least for energies greater than 20 eV, a factor of ~ 1.19 greater in magnitude than the measurement. This illustrates the value of incorporating Q space into the calculations through the optical potential. Nonetheless again we believe a major reason for this observed discrepancy between our CCO8 results and the experiment is due to the accuracy of our P-space target wave functions. The present CCO8 calculation used 32 quadrature points in our computations, while that of Zhou *et al.* [18] employed only 24. We therefore believe the present CCO8 calculation is more reliable and certainly better converged than that of the previous work [18].

In Fig. 3 we now plot the present experimental low-energy ICS results in more detail, where they are compared against the earlier data from Flynn *et al.* [17]. There is clearly excellent agreement between these two sets of results, both in terms of the magnitude of the ICS and in the shape of the energy dependence, over the entire common energy range. The overall energy resolution in both of these measurements was about 300 meV; hence it was not possible to test for the existence of any resonance structure in this energy regime. The present ICS data are also compared in Fig. 3 against the earlier four-state CC results of Scheibner *et al.* [16] and Msezane and Henry [15], as well as the current eight-state CC and CCO calculation results.

When allowance is made for the finite-energy resolution of the electron beam, the present ICS measurements are consistent with the theoretical prediction [16] of the very steep onset of the integral cross section at threshold. The four-state CC calculation of Msezane and Henry [15] predicts essentially constant ICS values from 6 to 10 eV of magnitude $9.63\pi a_0^2$. If cascades are ignored in the process then this theory superficially agrees somewhat better with the present experimental data (between 6–10 eV) than that from Scheibner *et al.* [16], although that agreement is probably a little fortuitous. The present CC and CCO results are plotted here as discrete points corresponding to the energies at which they were calculated. As expected the eight-state CC result significantly overestimates the magnitude of the experimental cross sections. On the other hand, the present eight-state CCO result, to within the uncertainty on the data, is in good accord with the available measurements. While this suggests it would be worthwhile to run the eight-state CCO calculations on a finer energy grid in this energy region, we simply do not have the computational resources to undertake such a task at this time. In addition there are other approaches available [29,30] that might be better suited to tackle the problem at these lower energies.

V. CONCLUSIONS

We have reported measurements and calculations for electron-impact excitation of the $4\ ^2P$ state in copper. At intermediate energies the present results were found to be in good accord with the earlier DCS-derived ICS from Ismail and Teubner [10], and at lower energies they were also seen to be in good agreement with the results from Flynn *et al.*

[17]. We therefore believe that the present experimental ICSs represent a reliable set for both kinetic modeling of the function of the CVL and testing scattering theory, from threshold up to 100 eV. Our eight-state CC theory consistently overestimated the magnitude of ICS over the energy range considered, by a factor of about 1.6. This is also the same factor by which our velocity form of the optical oscillator strength overestimates the accepted value for the $4\ ^2P$ state. Hence we conclude that our target description is a major factor in this observed discrepancy. While our eight-state CCO calculation is in better agreement with the measurements, it too suffers from a less than perfect target description. Very recently, relativistic forms of the CCC theory [29] and *B*-spline *R*-matrix approach [30] have been developed. We believe that their application to this scattering system would be profitable.

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